Facile collisional dissociation of N_2 on a Si(111)-7x7 surface at room temperature

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The industrial activation and transformation of N_2 to ammonia are carried out according to the well-known Haber–Bosch process, an old process invented about 100 years ago by F. Haber and C. Bosch. Hence, this reaction consumes about 2% of the world's energy supply and accounts for 1.44% of the CO2 global emission.[1] Indeed, the limiting step of this reaction is the dissociation of N_2, from the triple N≡N bond thereof, which is the strongest bond in diatomic molecules.

In our work, we demonstrate that the strong N_2 bond can be efficiently dissociated at low pressure and ambient temperature on a Si(111)-7x7 surface. The reaction was experimentally investigated by scanning tunneling microscopy and X-ray photoemission spectroscopy. Experimental and density functional theory results suggest that relatively low thermal energy collision of N_2 with the surface can facilitate electron transfer from the Si(111)-7x7 surface to the π^*-antibonding orbitals of N_2 that significantly weaken the N_2 bond. This facile N_2 triple bond dissociation on the surface leads to the formation of a Si_3N interface.

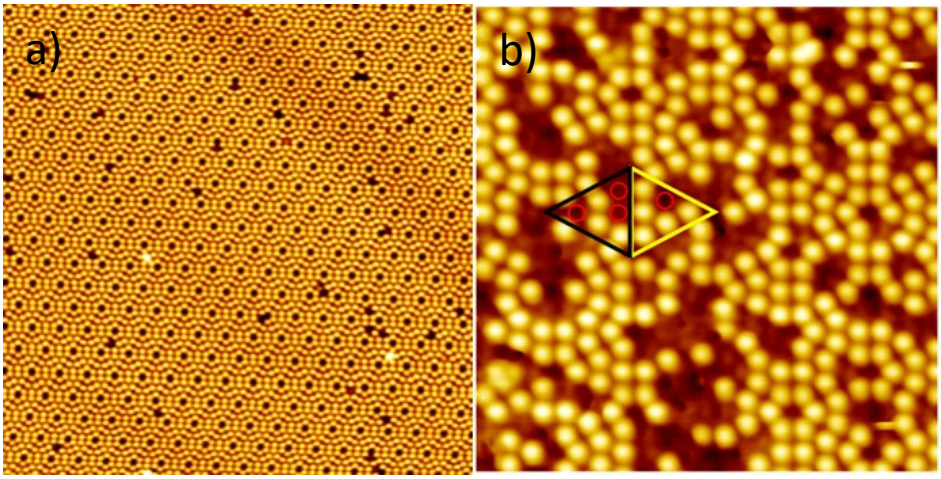

Figure 1: a) STM image (50x50 nm2, It = 200 pA, Vs = + 2 V) of the Si(111)-7x7 surface. b) STM image (15x15 nm^2, I_t = 200 pA, V_s = + 1.5 V) recorded after exposition at $1 \cdot 10^{-5}$ mbar for 30 minutes. Darker spots are attributed to modified silicon adatoms, both adatoms of faulted (yellow triangle) or unfaulted (black triangle) half-unit cells are affected.

Our results could be relevant for fine comprehension of the preliminary steps of the growth of silicon nitride, which is extremely important for microelectronics. The investigation of the ability of Si(111)-7x7 for efficient synthesis of ammonia is currently being investigated.

Références

[1] R. Schlogl, R. *Angew. Chem. Int. Ed.* **2003**, *42*, 2004-2008.